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Theoretical models and computational codes have been developed to describe chemical and physical phenomena associated with solids, microstructure clusters and polymers, with special attention given to nonlinear optical effects and ultrafast processes. The following topics have been investigated: light-induced drift of electrons in semiconductor heterostructures; photoinduced electron transfer in coupled quantum wells; quantum beats in time-resolved luminescence spectra; scale-invariant theory of optical properties of fractals; optical properties of small silicon clusters; boron-nitrogen-substituted fullerenes; and nonlinear optical response in polymers irradiated by laser fields.

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Contents

Summary

The objective under the current ONR grant was to develop theoretical models and computational codes to describe chemical and physical phenomena associated with solids, microstructure clusters and polymers, with special attention given to nonlinear optical effects and ultrafast processes. Several key results are summarized below.

- Light-Induced Drift of Electrons in Semiconductor Heterostructures. A novel effect of light-induced drift (LID) for quantum confined electrons has been predicted. This effect manifests itself as the electric current in the heterostructure plane in response to optical excitation with frequency close, but not exactly equal, to a resonance with an intersubband transition in the heterostructure. The current reverses its direction with a change in the detuning sign and vanishes if the radiation polarization is normal to the heterostructure plane. The LID effect is based upon the difference in relaxation times of an electron in different confined states. The current density has been estimated to be rather high, which makes the LID effect promising for applications in photonics.

- Photoinduced Electron Transfer in Coupled Quantum Wells. An effect of counterfield electron transfer has been predicted for an asymmetric double quantum well subjected to photoexcitation resonant with an intersubband electronic transition. The effect manifests itself in the transfer of electrons from one quantum well to the other well in the direction *opposite* to the one favored by the bias electric field. A quantitative theory of the effect has been developed on the basis of the density-matrix technique, which takes into account all types of relaxation. This technique has also been extended to describe the light-induced drift effect under optically saturated conditions. The theory shows that the counterfield electron transfer should be pronounced at realistic conditions and readily detectable. The transfer quantum yield is predicted to be high, up to 0.5 or greater. This effect is promising for use in far-infrared photodetection and in optoelectronic (photonic) devices, in particular, in photonic memory.

- Quantum Beats in Time-Resolved Luminescence Spectra. Using the above biased asymmetric double quantum well, analytical expressions have been developed for time-dependent luminescence intensities and numerically demonstrated to elucidate the characteristics of π-phase-shifted quantum beats. It is seen analytically as well as numerically that the magnitude of the tunneling interaction can be quantitatively estimated by the beat modulation depth.

- Scale-Invariant Theory of Optical Properties of Fractals. Surface-enhanced Raman scattering (SERS) from colloidal metal clusters, which are known to be fractal, is up to a millionfold greater in intensity than ordinary Raman scattering. Although there are some theories previously in the literature which qualitatively explain this magnitude of enhancement, none of them is capable of describing the spectral profiles of SERS. Under this ONR program a quantitative scale-invariant theory of SERS from fractals has been developed. The theory is supported by extensive numerical calculations with the use of supercomputers. The theory predicts scaling behavior of the SERS enhancement factor in terms of a properly chosen spectral variable. The theory also quantitatively describes the experimental spectral profiles of the SERS enhancement without a single adjustable parameter. The scale-invariant theory has also been extended to describe nonlinear optical polarizabilites of fractals.

- <u>Optical Properties of Small Silicon Clusters</u>. The geometry and electronic structure of small silicon clusters, Si_7, Si_{10} (three isomers) and Si_{13}, has been established using the *tight-binding model* by global optizimation of the cohesion energy. Expressions for linear and nonlinear polarizabilities of the clusters have been found using the single-particle density-matrix technique in the form of the sum-over-one-electron-states. Kleinman's conjecture for hyperpolarizabilities has been shown to be violated in the practically important frequency-degenerate case. Polarizabilities governing various optical phenomena, such as scattering and absorption, second-harmonic generation, optical rectification, nonlinear corrections to the refraction index, phase conjugation, etc., have been evaluated for a series of wavelengths. The linear absorption and hyperpolarizabilities have been shown to depend primarily on the symmetry of the clusters and only secondarily on their size. The hyperpolarizabilities prove to be high for the low-symmetry clusters.

- <u>Boron-Nitrogen-Substituted Fullerenes.</u> For the systems ($@B_2C_{58}$), ($@N_2C_{58}$), ($@BNC_{58}$), ($@C_{12}B_{24}N_{24}$) and ($@B_{30}N_{30}$), MNDO (modified neglect of differential overlap) calculations have been carried out for the heats of formation from benzene, naphthalene and their BN analogues, and it is found that all these hybrids are approximately as stable as buckminsterfullerene. Surprisingly, it is predicted that ($@B_{30}N_{30}$) is stable and should be relatively simple to synthesize from borazine.

- <u>Nonlinear Optical Response in Polymers Irradiated by Laser Fields.</u> A new phenomenon of splitting in the pump-probe spectrum of conjugated polymers has been found. Numerical results with parameters pertaining to a polydiacetylene-toluene-sulfonate (PTS) single crystal in an optical cavity have been obtained, yielding a spectrum of three dispersion structures: one is centered at the exciton resonance, and the other two are at one phonon frequency and two phonon frequencies below the resonance, respectively. Strong responses have been found around the threshold of the pump-field intensity for the occurrence of optical bistability. In addition, the transient pump-probe spectrum for a PTS single crystal has been calculated by solving numerically a set of Bloch-like optical differential equations. Phonon-induced excitonic bleaching is clearly shown. The above results are in qualitative agreement with experimental observations. Finally, surface-induced optical bistability has been found for a PTS chain near a metal surface. The bistability is accompanied by a reduced vacuum fluctuation.

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Publications (Technical Reports)

Each manuscript listed below corresponds by number to the Technical Report previously submitted to the Office of Naval Research for Grant N00014-90-J-1193. The major portion of these are referred journal articles, and the remainder are invited book chapters and conference proceedings.

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